

Supporting information

Determination of $^{87}\text{Sr}/^{86}\text{Sr}$ Ratios in Food Grains Characterized by Low Concentration Sr Using MC-ICP-MS

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Experimental: Reagents

Ultrapure analytical-grade (TAMAPURE-AA-100) HNO_3 , HCl , and HF purchased from Tama Chemicals Co., Ltd. (Kanagawa, Japan) and Milli-Q water with a resistivity of $>18.2 \text{ M}\Omega \cdot \text{cm}$ from Merck KGaA (Darmstadt, Germany) were used for all the experimental procedures in this study. For the sample preparation for the elemental analyses, single elemental 1000 mg kg^{-1} stock solutions of Sr, Rb, Ca, Fe, and Zn were purchased from Kanto Chemical Co., Inc. (Tokyo, Japan). As an internal standard element used for the elemental analyses, 1000 mg kg^{-1} stock solutions of rhodium (Rh) were purchased from FUJIFILM Wako Pure Chemical Corporation (Osaka, Japan).

Experimental: Labwares

The 30- and 7-mL perfluoroalkoxy alkane (PFA) vials with screw top lids (Savillex Corporation, MN, USA) were used for sample digestion, digested sample evaporation, purified Sr fraction collection, and sample preparation for the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio measurements using MC-ICP-MS. The 50-mL polypropylene bottles with screw top lids were used for the sample preparation for elemental analyses using ICP-MS. These labwares were cleaned prior to use with Milli-Q water and HNO_3 .

Experimental: Acid digestion of reference materials

Approximately 0.15 g of NIST SRM 1515 and NIST SRM 1570a as well as 0.5 g of NMIJ CRM 7502-a were weighed into separate polytetrafluoroethylene (PTFE) vessels. Additionally, ≈ 0.5 g of NIST SRM 1567a and NIST SRM 1567b were each placed in five separate PTFE vessels. Furthermore, to each PTFE vessel, 5 mL of a 25:1 mixture of 68 % HNO_3 and 38 % HF was added. The acid digestion of vegetal reference materials with only HNO_3 was incomplete; therefore, a small amount of HF was added to each vessel to dissolve the residual silicate material. Approximately 0.1 g of NIST SRM 1400, NIST SRM 1486, JcP-1, and FEBS-1 was weighted into separate PTFE vessels, and 5 mL of a 4:1 mixture of 68 % HNO_3 and 30 % HCl was added to each of these vessels, further ensuring the complete digestion of these reference materials were completely digested. By contrast, some residues remained in the digested solutions, and incomplete acid digestion occurred when only HNO_3 was used. The microwave program comprised three steps: temperature was linearly increased to 120 °C within 8 min; temperature was linearly increased to 230 °C within another 12 min; and temperature was kept at 230 °C for 20 min. Electric power was set at 1500 W over the entire three steps. The geological reference materials (JA1 and JB2) were digested via the hotplate digestion method. Approximately 0.1 g of JA1 and JB2 was weighted into a 30-mL PFA vial, and 5 mL of a 2:1 mixture of 68 % HNO_3 and 38 % HF was added to each vial. Closed vials were left on a hotplate at 130 °C for 48 h; further, the vials were opened and solutions were evaporated to dryness. To each vial, 5 mL of 30 % HCl was added and evaporated to dryness on the hotplate at 130 °C. This HCl addition and evaporation process were repeated thrice to eliminate fluorides, followed by adding 5 mL of 68 % HNO_3 .

Each digested sample solution was evaporated in a 30-mL PFA vial before Sr/matrix separation. The digested NIST SRM 1515, NIST SRM 1570a, NIST SRM 1400, NIST SRM 1486, JcP-1, and FEBS-1 were transferred from the PTFE digestion vessel to a 30-mL PFA vial. The geological reference materials digested using the hotplate

digestion method remained in their respective 30-mL PFA vials. NIST SRM 1567a and b contained particularly low Sr concentrations; hence, the digested solutions from five PTFE vessels prepared earlier (equivalent to 2.5 g powder) were collected into a PFA vial. Evaporation was performed on the hot plate at 130 °C until the volume of each solution was reduced to almost a single drop. Each drop from reference materials was redissolved in 2.5 mL 8 mol L⁻¹ HNO₃ and subjected to Sr/matrix separation.

Prior to determining Sr in reference materials via ICP-MS to validate the sample acid digestion method, the digested NIST SRM 1515, NIST SRM 1570a, NIST SRM 1400, NIST SRM 1486, NMIJ CRM 7502-a, and JCP-1 were transferred to individual 50-mL polypropylene bottles and diluted with an appropriate amount of 2 % HNO₃.

Experimental: Sample preparation for the elemental analyses

A 500-μL aliquot from each redissolved digested sample, purified Sr fraction, and procedural blank solution was diluted with an appropriate amount of 2 % (v/v) HNO₃ for the elemental analyses of Sr, Rb, Ca, Fe, and Zn using ICP-MS. The Sr concentration in each redissolved digested sample, purified Sr fraction, and blank solution was determined using the calibration method. As an internal standard element, 1 μg kg⁻¹ Rh was added to every measurement solution. The signal intensity of Rh originally contained in measurement solutions prepared from the redissolved digested sample and purified Sr fraction of each reference material was confirmed to be less than 0.5 % of the signal intensity of 1 μg kg⁻¹ Rh measured in advance.

Experimental: Blank correction, isobaric interference correction, and mass bias correction.

The mass bias was corrected based on the following equation:

$$\left(\frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}}\right)_{\text{corr}} = \left(\frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}}\right)_{\text{meas}} \times \left[\frac{\left(\frac{{}^{88}\text{Sr}}{{}^{86}\text{Sr}}\right)_{\text{ref}}}{\left(\frac{{}^{88}\text{Sr}}{{}^{86}\text{Sr}}\right)_{\text{meas}}} \right]^{\left[\frac{\ln\left(\frac{m87}{m86}\right)}{\ln\left(\frac{m88}{m86}\right)} \right]}$$

where “corr,” “meas,” and “ref” represent corrected, measured, and reference ratios, respectively. Here, “*m*” denotes the absolute mass for each isotope.

Tables

Table S1 Instrumental settings for ICP-MS (Agilent 7700x)

Instrument settings	
RF power (W)	1500
Argon gas flow rate (L min ⁻¹)	
Plasma	15
Makeup	0.4
Carrier	0.8
Axial sampling position (mm)	8
Sample introduction system	
Nebulizer	MicroMist
Spray chamber	Scott type
ICP Torch	Standard quartz torch with 2.5 mm ID injector, including a shield plate and a quartz bonnet
Sampling cone	Orifice diameter: 1 mm Pt
Skimmer cone	Orifice diameter: 0.4 mm Pt
Dwell time (ms)	10
Sweeps	100
Replicates	10
Isotopes measured	⁴⁴ Ca ⁺ , ⁵⁶ Fe ⁺ , ⁶⁶ Zn ⁺ , ⁸⁵ Rb ⁺ , ⁸⁸ Sr ⁺ , and ¹⁰³ Rh ⁺

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Vegetal						
NIST SRM 1515 (apple leaves)						
Digestion 1	C0.4-1	0.4	1.1	95.6	0.00012	
						Aliquot 1 0.71398
						Aliquot 2 0.71397
						Aliquot 3 0.71397
						Aliquot 4 0.71395
						Aliquot 5 0.71399
						Aliquot 6 0.71398
						Mean value \pm 2 SD 0.71397 \pm 0.00003 ($n = 6$)
Digestion 2	C0.4-2	0.4	1.2	99.9	0.00040	
						Aliquot 1 0.71399
						Aliquot 2 0.71398
						Aliquot 3 0.71397
						Aliquot 4 0.71397
						Aliquot 5 0.71398
						Mean value \pm 2 SD 0.71398 \pm 0.00002 ($n = 5$)
Digestion 3	C0.4-3	0.4	0.9	100.1	0.00095	
						Aliquot 1 0.71398
						Aliquot 2 0.71399
						Aliquot 3 0.71399
						Mean value \pm 2 SD 0.71399 \pm 0.00001 ($n = 3$)
Digestion 4	C0.4-4	0.4	1.1	96.1	0.00100	
						Aliquot 1 0.71397
						Aliquot 2 0.71395
						Aliquot 3 0.71396
						Mean value \pm 2 SD 0.71396 \pm 0.00003 ($n = 3$)
Digestion 5	C2.0-1	2.0	2.1	95.4	0.00013	
						Aliquot 1 0.71395
						Aliquot 2 0.71398
						Aliquot 3 0.71397
						Aliquot 4 0.71398
						Mean value \pm 2 SD 0.71397 \pm 0.00002 ($n = 4$)
						Mean value \pm 2 SD 0.71397 \pm 0.00003 ($n = 21$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using

MC-ICP-MS.

Table S3 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Vegetal						
NIST SRM 1570a (spinach)						
Digestion 1	C0.4-5	0.4	1.3	98.2	0.00034	
						Aliquot 1 0.70906
						Aliquot 2 0.70903
						Aliquot 3 0.70904
						Aliquot 4 0.70904
						Mean value \pm 2 SD 0.70904 \pm 0.00003 ($n = 4$)
Digestion 2	C0.4-6	0.4	1.8	96.4	0.00035	
						Aliquot 1 0.70906
						Aliquot 2 0.70904
						Aliquot 3 0.70904
						Aliquot 4 0.70905
						Mean value \pm 2 SD 0.70905 \pm 0.00002 ($n = 4$)
Digestion 3	C0.4-7	0.4	1.9	95.5	0.00100	
						Aliquot 1 0.70904
						Aliquot 2 0.70905
						Aliquot 3 0.70904
						Aliquot 4 0.70903
						Mean value \pm 2 SD 0.70904 \pm 0.00001 ($n = 4$)
Digestion 4	C0.4-8	0.4	2.6	98.1	0.00098	
						Aliquot 1 0.70905
						Aliquot 2 0.70906
						Aliquot 3 0.70907
						Mean value \pm 2 SD 0.70906 \pm 0.00002 ($n = 3$)
Digestion 5	C2.0-2	2.0	2.6	98.4	0.00015	
						Aliquot 1 0.70903
						Aliquot 2 0.70904
						Aliquot 3 0.70905
						Aliquot 4 0.70906
						Aliquot 5 0.70904
						Mean value \pm 2 SD 0.70905 \pm 0.00002 ($n = 5$)
						Mean value \pm 2 SD 0.70905 \pm 0.00002 ($n = 20$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Animal						
NIST SRM 1400 (bone ash)						
Digestion 1	C0.4-9	0.4	1.7	98.8	0.00029	
						Aliquot 1 0.71313
						Aliquot 2 0.71312
						Aliquot 3 0.71313
						Mean value \pm 2 SD 0.71312 \pm 0.00001 ($n = 3$)
Digestion 2	C0.4-10	0.4	1.7	95.7	0.00005	
						Aliquot 1 0.71313
						Aliquot 2 0.71313
						Aliquot 3 0.71312
						Aliquot 4 0.71313
						Aliquot 5 0.71314
						Mean value \pm 2 SD 0.71313 \pm 0.00002 ($n = 5$)
Digestion 3	C2.0-3	2.0	4.5	95.8	0.00003	
						Aliquot 1 0.71313
						Aliquot 2 0.71312
						Aliquot 3 0.71313
						Mean value \pm 2 SD 0.71312 \pm 0.00001 ($n = 3$)
Digestion 4	C2.0-4	2.0	4.5	97.2	0.00005	
						Aliquot 1 0.71312
						Aliquot 2 0.71312
						Aliquot 3 0.71313
						Mean value \pm 2 SD 0.71313 \pm 0.00002 ($n = 3$)
Digestion 5	C2.0-5	2.0	4.5	98.4	0.00003	
						Aliquot 1 0.71314
						Aliquot 2 0.71313
						Aliquot 3 0.71313
						Mean value \pm 2 SD 0.71313 \pm 0.00001 ($n = 3$)
						Mean value \pm 2 SD 0.71313 \pm 0.00001 ($n = 17$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Animal						
NIST SRM 1486 (bone meal)						
Digestion 1	C0.4-11	0.4	1.7	98.8	0.00009	
						Aliquot 1 0.70933
						Aliquot 2 0.70932
						Aliquot 3 0.70932
						Aliquot 4 0.70934
						Mean value \pm 2 SD 0.70932 \pm 0.00001 ($n = 4$)
Digestion 2	C0.4-12	0.4	1.7	99.3	0.00011	
						Aliquot 1 0.70933
						Aliquot 2 0.70930
						Aliquot 3 0.70931
						Aliquot 4 0.70932
						Mean value \pm 2 SD 0.70931 \pm 0.00003 ($n = 4$)
Digestion 3	C2.0-6	2.0	4.3	95.7	0.00004	
						Aliquot 1 0.70930
						Aliquot 2 0.70928
						Aliquot 3 0.70930
						Aliquot 4 0.70931
						Mean value \pm 2 SD 0.70930 \pm 0.00003 ($n = 4$)
Digestion 4	C2.0-7	2.0	4.3	98.4	0.00004	
						Aliquot 1 0.70929
						Aliquot 2 0.70930
						Aliquot 3 0.70929
						Aliquot 4 0.70932
						Mean value \pm 2 SD 0.70930 \pm 0.00003 ($n = 4$)
Digestion 5	C2.0-8	2.0	4.4	96.5	0.00003	
						Aliquot 1 0.70929
						Aliquot 2 0.70932
						Aliquot 3 0.70931
						Mean value \pm 2 SD 0.70931 \pm 0.00003 ($n = 3$)
						Mean value \pm 2 SD 0.70931 \pm 0.00003 ($n = 19$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Animal						
GSJ JCP-1 (coral)						
Digestion 1	C0.4-13	0.4	13.2	95.6	0.00007	
						Aliquot 1 0.70918
						Aliquot 2 0.70918
						Aliquot 3 0.70917
						Mean value \pm 2 SD 0.70918 \pm 0.00001 ($n = 3$)
Digestion 2	C2.0-9	2.0	15.3	97.2	0.00007	
						Aliquot 1 0.70918
						Aliquot 2 0.70916
						Aliquot 3 0.70915
						Aliquot 4 0.70918
						Aliquot 5 0.70917
						Mean value \pm 2 SD 0.70917 \pm 0.00001 ($n = 5$)
Digestion 3	C2.0-10	2.0	29.4	97.4	0.00004	
						Aliquot 1 0.70916
						Aliquot 2 0.70915
						Aliquot 3 0.70917
						Aliquot 4 0.70917
						Mean value \pm 2 SD 0.70916 \pm 0.00001 ($n = 4$)
Digestion 4	C2.0-11	2.0	29.5	98.4	0.00003	
						Aliquot 1 0.70917
						Aliquot 2 0.70917
						Aliquot 3 0.70918
						Mean value \pm 2 SD 0.70917 \pm 0.00001 ($n = 3$)
Digestion 5	C2.0-12	2.0	29.3	96.5	0.00004	
						Aliquot 1 0.70917
						Aliquot 2 0.70918
						Aliquot 3 0.70917
						Mean value \pm 2 SD 0.70917 \pm 0.00001 ($n = 3$)
						Mean value \pm 2 SD 0.70917 \pm 0.00002 ($n = 18$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
Animal						
NRC FEBS-1 (otolith)						
Digestion 1	C0.4-14	0.4	12.8	98.4	0.00005	
						Aliquot 1 0.70916
						Aliquot 2 0.70917
						Aliquot 3 0.70918
						Mean value \pm 2 SD 0.70917 \pm 0.00001 ($n = 3$)
Digestion 2	C0.4-15	0.4	12.9	95.8	0.00004	
						Aliquot 1 0.70917
						Aliquot 2 0.70918
						Aliquot 3 0.70918
						Mean value \pm 2 SD 0.70918 \pm 0.00002 ($n = 3$)
Digestion 3	C2.0-13	2.0	12.6	98.3	0.00007	
						Aliquot 1 0.70918
						Aliquot 2 0.70918
						Aliquot 3 0.70918
						Aliquot 4 0.70918
						Mean value \pm 2 SD 0.70918 \pm 0.00001 ($n = 4$)
Digestion 4	C2.0-14	2.0	12.7	97.5	0.00003	
						Aliquot 1 0.70918
						Aliquot 2 0.70918
						Aliquot 3 0.70917
						Aliquot 4 0.70918
						Aliquot 5 0.70916
						Mean value \pm 2 SD 0.70917 \pm 0.00001 ($n = 5$)
Digestion 5	C2.0-15	2.0	12.7	96.8	0.00005	
						Aliquot 1 0.70918
						Aliquot 2 0.70917
						Aliquot 3 0.70918
						Aliquot 4 0.70917
						Aliquot 5 0.70916
						Mean value \pm 2 SD 0.70917 \pm 0.00002 ($n = 5$)
						Mean value \pm 2 SD 0.70917 \pm 0.00002 ($n = 20$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$	
Geological						
GSJ JA1 (andesite)						
Digestion 1	C0.4-16	0.4	4.7	95.5	0.00017	
					Aliquot 1	0.70355
					Aliquot 2	0.70356
					Aliquot 3	0.70355
					Aliquot 4	0.70355
					Mean value \pm 2 SD	0.70355 \pm 0.00002 ($n = 4$)
Digestion 2	C2.0-16	2.0	2.4	93.4	0.00020	
					Aliquot 1	0.70354
					Aliquot 2	0.70354
					Aliquot 3	0.70353
					Aliquot 4	0.70355
					Mean value \pm 2 SD	0.70354 \pm 0.00002 ($n = 4$)
					Mean value \pm 2 SD	0.70355 \pm 0.00002 ($n = 10$)
Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$	
GSJ JB2 (basalt)						
Digestion 1	C0.4-17	0.4	3.5	90.8	0.00013	
					Aliquot 1	0.70369
					Aliquot 2	0.70369
					Aliquot 3	0.70369
					Aliquot 4	0.70369
					Aliquot 5	0.70367
					Mean value \pm 2 SD	0.70369 \pm 0.00002 ($n = 5$)
Digestion 2	C2.0-17	2.0	3.4	91.2	0.00012	
					Aliquot 1	0.70367
					Aliquot 2	0.70369
					Aliquot 3	0.70368
					Aliquot 4	0.70366
					Aliquot 5	0.70367
					Aliquot 6	0.70368
					Mean value \pm 2SD	0.70368 \pm 0.00002 ($n = 6$)
					Mean value \pm 2SD	0.70368 \pm 0.00002 ($n = 11$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)	$^{87}\text{Sr}/^{86}\text{Sr}$
NIST SRM 1567a (wheat flour)						
Digestion 1	C0.4-18	0.4	1.8	97.8	0.00027	
						Aliquot 1 0.70893
						Aliquot 2 0.70894
						Aliquot 3 0.70893
						Aliquot 4 0.70897
						Aliquot 5 0.70892
						Mean value \pm 2 SD 0.70894 \pm 0.00004 ($n = 5$)
Digestion 2	C0.4-19	0.4	2.0	98.1	0.00016	
						Aliquot 1 0.70892
						Aliquot 2 0.70893
						Aliquot 3 0.70893
						Aliquot 4 0.70892
						Aliquot 5 0.70892
						Aliquot 6 0.70893
						Aliquot 7 0.70896
						Mean value \pm 2 SD 0.70893 \pm 0.00003 ($n = 6$)
Digestion 3	C0.4-20	0.4	1.9	96.4	0.00019	
						Aliquot 1 0.70898
						Aliquot 2 0.70899
						Aliquot 3 0.70895
						Mean value \pm 2 SD 0.70898 \pm 0.00004 ($n = 3$)
Digestion 4	C2.0-18	0.4	1.9	97.4	0.00016	
						Aliquot 1 0.70899
						Aliquot 2 0.70899
						Aliquot 3 0.70897
						Aliquot 4 0.70896
						Aliquot 5 0.70897
						Aliquot 6 0.70895
						Aliquot 7 0.70899
						Mean value \pm 2 SD 0.70898 \pm 0.00003 ($n = 7$)
Digestion 5	C2.0-19	0.4	2.0	95.2	0.00007	
						Aliquot 1 0.70894
						Aliquot 2 0.70897
						Aliquot 3 0.70895
						Aliquot 4 0.70898
						Mean value \pm 2 SD 0.70896 \pm 0.00004 ($n = 4$)
Digestion 6	C2.0-20	2.0	2.0	95.7	0.00024	
						Aliquot 1 0.70893
						Aliquot 2 0.70892
						Aliquot 3 0.70893
						Aliquot 4 0.70895
						Aliquot 5 0.70892
						Mean value \pm 2 SD 0.70893 \pm 0.00003 ($n = 5$)
Digestion 7	C2.0-21	2.0	2.0	98.5	0.00010	
						Aliquot 1 0.70892
						Aliquot 2 0.70893
						Aliquot 3 0.70896
						Aliquot 4 0.70893
						Mean value \pm 2 SD 0.70893 \pm 0.00003 ($n = 4$)
						Mean value \pm 2 SD 0.70895 \pm 0.00005 ($n = 34$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed

using MC-ICP-MS.

Table S2 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for reference materials measured in this study

	Column name	Sr resin bed volume (mL)	Sr mass loaded (μg)	Sr recovery (%)	$^{85}\text{Rb}/^{88}\text{Sr}^*$ (%)		$^{87}\text{Sr}/^{86}\text{Sr}$
NIST SRM 1567b (wheat flour)							
Digestion 1	C0.4-21	2.0	2.0	98.3	0.00014		
						Aliquot 1	0.70895
						Aliquot 2	0.70896
						Aliquot 3	0.70896
						Aliquot 4	0.70897
						Aliquot 5	0.70899
						Mean value \pm 2 SD	0.70897 \pm 0.00003 ($n = 4$)
Digestion 2	C0.4-22	2.0	2.0	95.5	0.00013		
						Aliquot 1	0.70896
						Aliquot 2	0.70897
						Aliquot 3	0.70899
						Mean value \pm 2 SD	0.70897 \pm 0.00002 ($n = 3$)
Digestion 3	C0.4-23	2.0	2.0	96.7	0.00015		
						Aliquot 1	0.70899
						Aliquot 2	0.70897
						Aliquot 3	0.70898
						Mean value \pm 2 SD	0.70898 \pm 0.00002 ($n = 3$)
Digestion 4	C0.4-24	0.4	2.0	98.2	0.00019		
						Aliquot 1	0.70900
						Aliquot 2	0.70900
						Aliquot 3	0.70901
						Aliquot 4	0.70900
						Aliquot 5	0.70898
						Aliquot 6	0.70901
						Mean value \pm 2 SD	0.70900 \pm 0.00001 ($n = 6$)
Digestion 5	C0.4-25	0.4	2.0	97.5	0.00013		
						Aliquot 1	0.70897
						Aliquot 2	0.70899
						Aliquot 3	0.70899
						Aliquot 4	0.70898
						Mean value \pm 2 SD	0.70898 \pm 0.00001 ($n = 4$)
Digestion 6	C2.0-22	0.4	2.0	97.6	0.00011		
						Aliquot 1	0.70897
						Aliquot 2	0.70897
						Aliquot 3	0.70899
						Aliquot 4	0.70899
						Aliquot 5	0.70899
						Mean value \pm 2 SD	0.70898 \pm 0.00001 ($n = 5$)
						Mean value \pm 2 SD	0.70898 \pm 0.00003 ($n = 25$)

*Each value indicates the signal intensity ratio of $^{85}\text{Rb}/^{88}\text{Sr} \times 100$ obtained when each diluted Sr fraction is analyzed using

MC-ICP-MS.

Table S3. Comparison of the measured and certified values of the Sr concentrations

	Measured value ^{a)} (mg kg ⁻¹)			Certified value ^{b)} (mg kg ⁻¹)		
Vegetal						
NIST SRM 1515 (apple leaves)	25.5	±	0.06	25.1	±	1.1
NMIJ CRM 7502-a (rice flour) ^{c)}	0.068	±	0.002	0.068	±	0.003
NIST SRM 1570a (spinach)	55.46	±	0.21	55.54	±	0.50
Animal						
NIST SRM 1400 (bone ash)	245	±	2	249	±	7
NIST SRM 1486 (bone meal)	260	±	2	264	±	7
GSJ JCP-1 (coral)	7239	±	34	7240	±	70

^{a)}Each value represents the mean value ± standard uncertainty. Each standard uncertainty is estimated by combining the relative standard uncertainty of the analysis and Sr standard solution used for calibration. ^{b)}Each value denotes the property value ± expanded uncertainty (95 % confidence interval). ^{c)}The Sr mass fractions of NIST SRM 1567a and b are not certified; thus, the acid digestion method of NIST SRM 1567a and b is validated using a reference material of rice flour, which has a similar sample matrix to wheat flour.

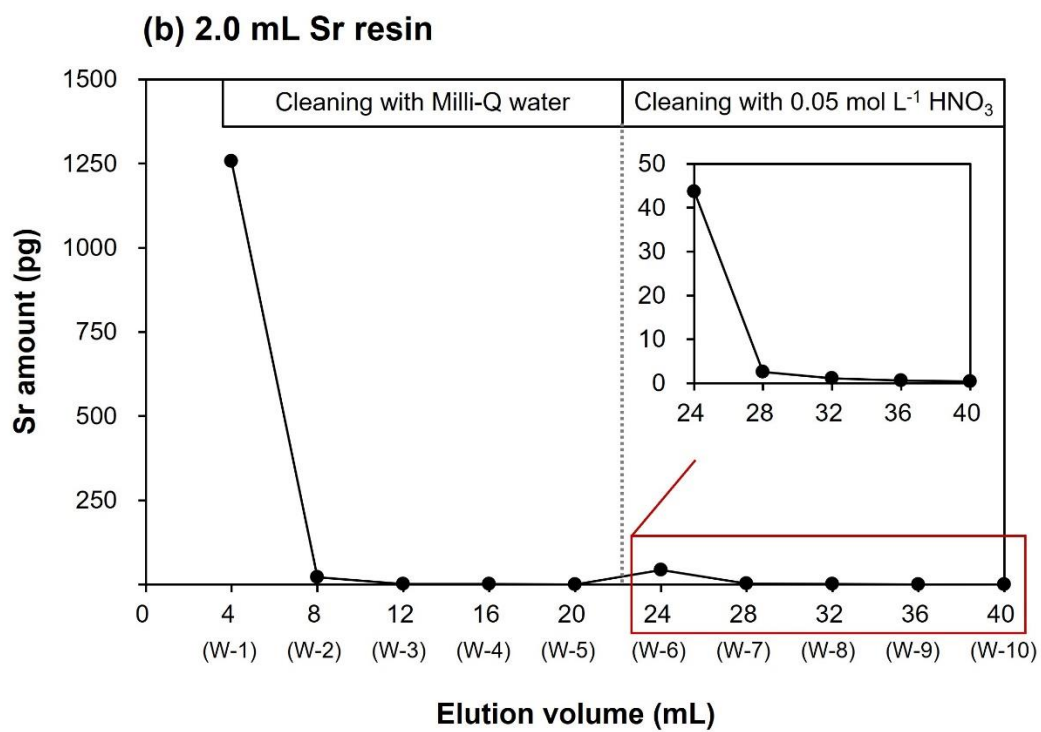
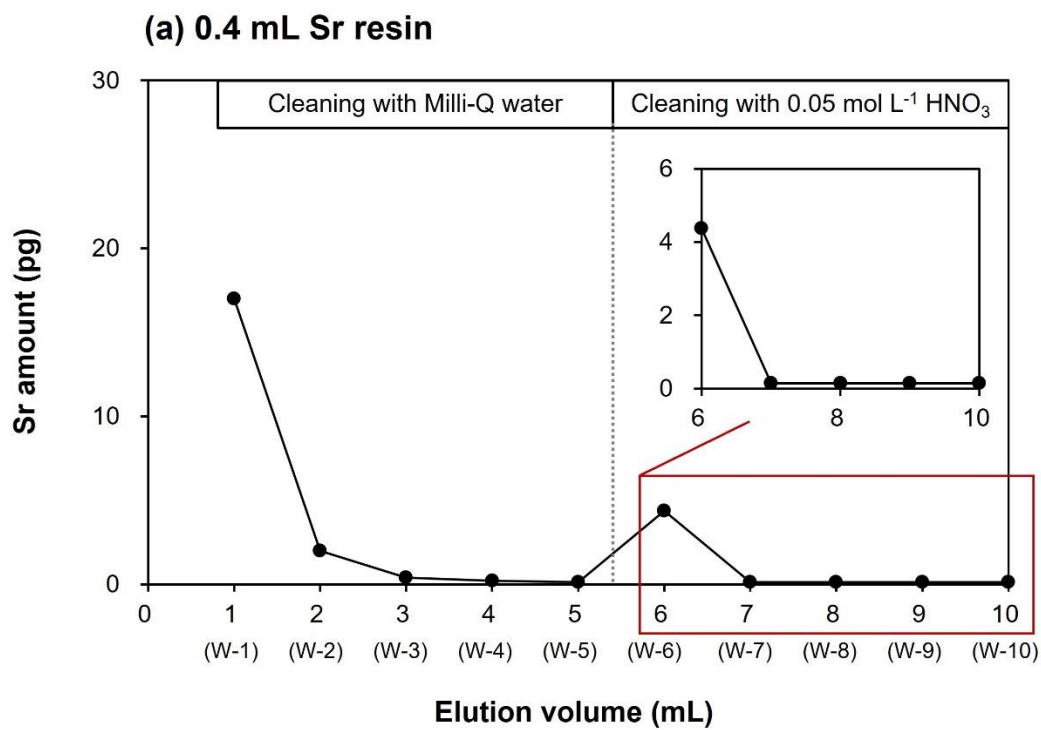


Fig. S1 Elution profile of Sr during the resin cleaning step for 0.4 mL (a) and 2.0 mL (b) Sr resins. Inset shows the elution profile in the range of 6–10 mL (a) and 24–40 mL (b).